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23-07-2015	Final Report	6-Aug-2012 - 5-Aug-2013

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Final Report: Tunable ultrafast photon source and imaging system for studying carrier dynamics in graphene devices

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W911NF-12-1-0345

5b. GRANT NUMBER

5c. PROGRAM ELEMENT NUMBER
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6. AUTHORS

Brian LeRoy, Arvinder Sandhu

5d. PROJECT NUMBER

5e. TASK NUMBER

5f. WORK UNIT NUMBER

7. PERFORMING ORGANIZATION NAMES AND ADDRESSES

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14. ABSTRACT

This project enabled the acquisition of a optical parametric amplifier operating at energies ranging from 0.75-5 eV. In addition it allowed the purchase of a CCD camera operating in the near-IR wavelength range. This equipment is being used to study carrier dynamics in graphene and other two-dimensional materials.

15. SUBJECT TERMS

Graphene, Optical Parametric Amplifier, Raman Spectroscopy

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	15. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a. REPORT	b. ABSTRACT	c. THIS PAGE			Brian LeRoy
UU	UU	UU	UU		19b. TELEPHONE NUMBER 520-626-4726

Report Title

Final Report: Tunable ultrafast photon source and imaging system for studying carrier dynamics in graphene devices

ABSTRACT

This project enabled the acquisition of a optical parametric amplifier operating at energies ranging from 0.75-5 eV. In addition it allowed the purchase of a CCD camera operating in the near-IR wavelength range. This equipment is being used to study carrier dynamics in graphene and other two-dimensional materials.

Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

Received	Paper

TOTAL:

Number of Papers published in peer-reviewed journals:

(b) Papers published in non-peer-reviewed journals (N/A for none)

Received	Paper

TOTAL:

Number of Papers published in non peer-reviewed journals:

(c) Presentations

"Ultrafast pump-probe spectroscopy and microscopy of graphene and graphene heterostructures", Gordon Research Conference on Ultrafast Phenomena in Cooperative Systems, Ventura CA, Feb 2-7, 2014 (Talk given by Dheeraj Golla, graduate student).
"Imaging and spectroscopy of graphene heterostructures," March Meeting of the American Physical Society, Denver CO, March 2014 (Talk given by Brian LeRoy, PI).
"Gated Raman spectroscopy of twisted bilayer graphene," March Meeting of the American Physical Society, Denver CO, March 2014 (Talk given by Shengqiang Huang, graduate student).

Number of Presentations: 3.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received	Paper

TOTAL:

Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received	Paper

TOTAL:

Number of Peer-Reviewed Conference Proceeding publications (other than abstracts):

(d) Manuscripts

Received	Paper	
07/23/2015	1.00	Shengqiang Huang, Matthew Yankowitz, Kanokporn Chattrakun, Arvinder Sandhu, Brian J. LeRoy. Evolution of the electronic band structure of twisted bilayer graphene upon doping, SUBMITTED (05 2015)

TOTAL: **1**

Number of Manuscripts:

Books

Received Book

TOTAL:

Received Book Chapter

TOTAL:

Patents Submitted

Patents Awarded

Awards

Graduate Students

NAME	PERCENT_SUPPORTED	Discipline
Dheeraj Golla	0.00	
Shengqiang Huang	0.00	
FTE Equivalent:	**0.00**	
Total Number:	**2**	

Names of Post Doctorates

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Names of Faculty Supported

NAME	PERCENT_SUPPORTED	National Academy Member
Brian LeRoy	0.00	
Arvinder Sandhu	0.00	
FTE Equivalent:	**0.00**	
Total Number:	**2**	

Names of Under Graduate students supported

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Student Metrics

This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period: 0.00

The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:..... 0.00

The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:..... 0.00

Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):..... 0.00

Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering:..... 0.00

The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense 0.00

The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields:..... 0.00

Names of Personnel receiving masters degrees

NAME
Total Number:

Names of personnel receiving PHDs

NAME
Total Number:

Names of other research staff

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Sub Contractors (DD882)

Inventions (DD882)

Scientific Progress

See Attachment

Technology Transfer

Tunable ultrafast photon source and imaging system for studying carrier dynamics in graphene devices

As discussed below the focus of this DURIP project was on understanding the interaction between electrons, holes and phonons in graphene through a combination of two optical techniques – tunable wavelength ultrafast optical spectroscopy and Raman spectroscopy. Ultrafast pulse excitations create non-equilibrium conditions between various excitations, allowing coupling mechanisms and relaxation rates to be probed as a function of time. The structural properties of graphene devices were probed through scanning probe microscopy and Raman spectroscopy. Scanning probe microscopy provides direct images of the morphology of the samples while Raman spectroscopy gives access to the vibrational modes of the system. In the following we discuss the utilization of funds for acquiring the equipment which forms the basis for ongoing research on the opto-electronic properties of two-dimensional materials, including graphene and its heterostructures.

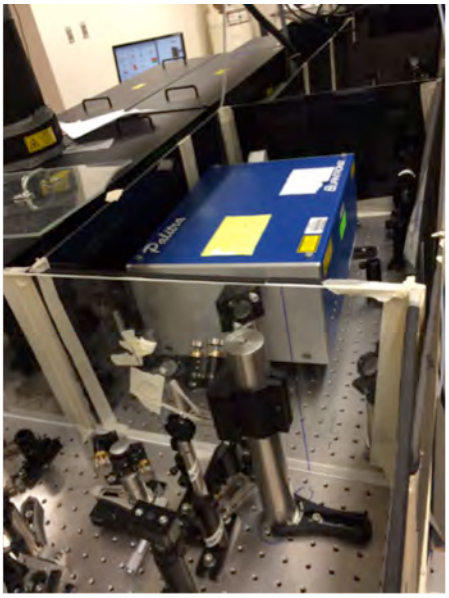

Fig 1. The set up for ultrafast spectroscopy showing the Quantronix OPA, Model: Palitra, (blue box) interfaced with femtosecond laser system and the station for ultrafast pump-probe experimental studies.

New instrumentation enabled by the DURIP funds:

Our ultrafast measurement capabilities were substantially enhanced through the acquisition of an Optical Parametric Amplifier (OPA) which was added-on to our existing femtosecond laser system. The use of OPA has opened avenues for study of the electron excitation and relaxation dynamics from 0.75 - 5 eV, allowing probing of the interesting physics near the saddle point of graphene band structure. In addition, this energy range will allow a wide range of transition metal dichalcogenides which have bandgaps ranging from 0.8 – 2 eV to be probed. Figure 1 shows the pump-probe ultrafast spectroscopy set up, where the OPA (blue box) is prominently shown next to the femtosecond laser system.

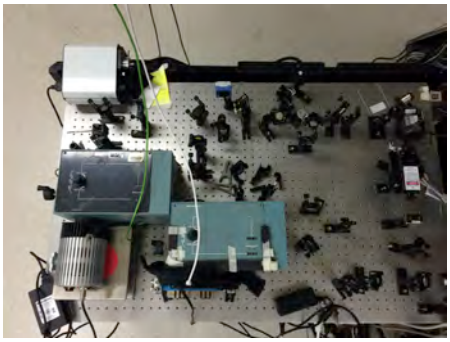

Fig 2. The set up for Raman spectroscopy showing the NIR CCD camera (BaySpec) in the bottom left corner.

We upgraded our Raman microscope through the acquisition of a TE-cooled CCD camera in the near-IR wavelength range. This is allowing us to study the phonon properties of 2D materials at lower energies (<1 eV), where we can probe the electron phonon interactions near the superlattice Dirac points created in bilayer and hetero-structure systems (for example of graphene and boron nitride). Figure 2 shows the homebuilt Raman spectroscopy setup in our lab which uses two wavelengths, 532 nm and 1064 nm. The near-infrared line array CCD camera for 1064 nm operation was

purchased from BaySpec using the DURIP funds. This setup is vital for characterization of all two dimensional samples prepared in our labs.

Experimental work:

We are pursuing our existing line of investigation to understand the many-body interactions in graphene. This work is a collaboration between PIs Brian LeRoy and Arvinder Sandhu in the physics department at the University of Arizona, as well as Henry Everitt at the Army AMRDEC in Huntsville, AL, with theoretical support being lent by Prof. Rolf Binder in the College of Optical Sciences at the University of Arizona.

We subjected graphene to an ultrashort laser pulse at 4.7 eV, and observed its response over a wide range of energies using a delayed probe pulse. Through a differential measurement of the transmission of the probe, we have observed a dynamic band structure modification caused by the photo-excited carriers and the subsequently generated phonons. These measurements allow us to quantify the many-body effects such as electron-phonon coupling in non-equilibrium graphene. In the ongoing measurements, we are extending this approach to the study of graphene-hBN interfaces, where we expect new effects due to the presence of pseudo-Dirac points in the band structure. In a run up to these studies, we have also developed a technique to optically image boron nitride flakes with precise thickness determination.

Using the Raman spectroscopy setup we have investigated, the electronic band structure of twisted bilayer graphene, and how it depends on the twist angle between the two layers. We observed an enhanced Raman G peak when the excitation laser is resonant with the energy separation of the van Hove singularities in the density of states where the two sets of monolayer bands cross (Fig 3). The variation of the Raman G peak area with charge density reveals doping induced changes in the electronic band structure. We have recently submitted a manuscript discussing these new results.

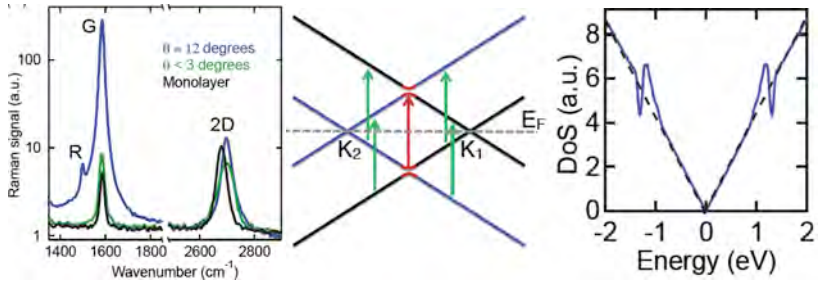

Fig 3. The observed enhancement in Raman G peak of bilayer graphene when the excitation resonantly couples to the van Hove singularities in the density of states due to crossing of monolayer graphene bands.

Research Products:

The following publications were an indirect byproduct of the activities supported by this project. The submitted paper and ongoing research are however directly associated with use of DURIP enabled instrumentation.

1) Adam Roberts, Rolf Binder, Nai-Hang Kwong, Dheeraj Golla, Daniel Cormode, Brian LeRoy, Henry Everitt, Arvinder Sandhu, "Optical characterization of electron-phonon interactions at the saddle point in graphene", *Phys. Rev. Lett.* **112**, 187401 (2014).

2) Dheeraj Golla, K. Chattrakun, K. Watanabe, T. Taniguchi, Brian J. LeRoy, Arvinder Sandhu, “Optical thickness determination of hexagonal boron nitride flakes” *Appl. Phys. Lett.* **102**, 161906 (2013).
3) Shengqiang Huang, Matthew Yankowitz, Kanokporn Chattrakun, Arvinder Sandhu, Brian J. LeRoy, “Evolution of the electronic band structure of twisted bilayer graphene upon doping” (**Submitted**)

Human Resource Development:

The instrumentation purchased using this grant is enabling the Ph.D. research of three graduate students, Alexandra Brasington, Dheeraj Golla, and Shengqiang Huang.